Energy relaxation dynamics in vertically coupled multi-stacked InAs/GaAs quantum dots

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Effect of GaAs spacer layer thickness (d_{GaAs}) on carrier capture, and the relaxation process is studied in multi-stacked InAs/GaAs quantum dots by photoluminescence and time-resolved photoluminescence. Auger scattering is the dominating process for carrier relaxation above d_{GaAs} of 15 nm. At d_{GaAs} of 10 nm, the carrier relaxation process is faster due to the combined effect of both single longitudinal optical phonon and Auger scattering resulting in higher photoluminescence intensity. The photoluminescence rise time corresponding to carrier capture and relaxation in quantum dots is longer at 3.06 eV excitation than that at 1.53 eV due to the effect of intervalley scattering in GaAs. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4974221>]

Quantum dots (QDs) have been the potential candidates for different electronic and optoelectronic devices such as lasers,¹ light emitting diodes,² infrared detectors,³ single electron transistors,⁴ etc. Recently, InAs/GaAs based QDs self-assembled in the Stranski-Krastanov (SK) growth mode have been viewed with significant interest.^{1,5} Devices based on such QDs operate at wavelengths as long as 1.2 μm , and there is a great interest in extending this wavelength to 1.55 μm and longer for use in fiber optic telecommunication systems.⁵ Multi-stacked (MS) InAs QDs with GaAs spacer have been the very promising device structures for the enhancement of the modal gain and the radiative efficiency.^{6,7} The GaAs spacer layer thickness (d_{GaAs}) has been reported to have a significant influence on the structural and optical properties of MS InAs/GaAs QDs.^{8–11} At larger d_{GaAs} , QDs are not vertically correlated, and each QD layer exhibits properties similar to that of a single layer. However, properties of MS QDs are different from the single QDs layer for smaller d_{GaAs} due to the vertical correlation of QDs.⁹ Energy relaxation process in single layer self-assembled InAs/GaAs QDs have widely been reported.^{12–15} The intra-dot relaxation process has been expected to be slower than the radiative recombination process (several nanoseconds) due to the atomic-like discrete eigen states of QDs with energy spacing much larger than the longitudinal optical (LO) phonon energy, which is known as the “phonon bottleneck” effect.¹⁶ Experimental studies have revealed a fast intra-dot carrier relaxation (<80 ps) which is in contrast to the phonon bottleneck effect.^{12–14} Recently, Kim *et al.*¹⁷ have observed the dominance of excited state emission in the photoluminescence (PL) spectra of MS InAs/GaAsSb QDs at and above the spacer layer thickness of 10 nm. They have also reported that the increase in the PL intensity ratio of

excited state emission to ground state emission with increasing spacer layer thickness is attributed to the slower carrier relaxation rate which is in accordance with the phonon bottleneck effect. However, systematic investigation on the carrier relaxation process at different spacer layer thicknesses in InAs based MS QDs has not been reported which is inevitable to make them suitable for various electronic and optoelectronics applications.

In this letter, we have discussed the PL and time-resolved photoluminescence (TRPL) properties of MS InAs/GaAs QDs at different d_{GaAs} . PL peak energy is found to be increased monotonically with decreasing d_{GaAs} which is in contrast to the earlier reports.^{8–11,17} A standard TRPL upconversion apparatus is used to perform an extensive study on the carrier dynamics and the intervalley scattering in multi-stacked InAs/GaAs QDs. Single LO phonon scattering along with Auger scattering process is found to be responsible for the faster carrier relaxation at reduced d_{GaAs} (10 nm).

The self-assembled MS InAs/GaAs QDs were grown by molecular beam epitaxy in the SK growth mode on (100)-oriented n-type GaAs substrate. A buffer layer of 0.5 μm thick GaAs, and 30 nm thick AlAs and 60 nm thick GaAs barrier layers were grown before 30 cycles of 2.6 monolayers of InAs with GaAs spacer layers were deposited. In order to cover the surface, 60 nm thick GaAs and 30 nm thick AlAs barrier layers, and then 5 nm GaAs cap layers were deposited.

MS InAs/GaAs QDs were characterized by transmission electron microscopy (TEM), atomic force microscopy (AFM), PL, and TRPL. AFM analysis reveals that the surface density of QDs in the investigated samples is about $5 \times 10^{10} \text{ cm}^{-2}$. The vertical height of the QDs is about 6–7 nm and the width of 20–30 nm. The PL measurements were performed at 35 K using a He closed-cycle cryostat. Frequency doubled (3.06 eV) output of a mode-locked Ti: Sapphire laser (pulse width ~ 100 fs, repetition rate = 80 MHz) were used as the excitation source. TRPL was measured at different photon energies (E_p) of the PL band at 35 K. The experimental configuration is similar to that in the previous reports.^{18,19}

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FIG. 1. Cross-sectional TEM images of the MS InAs/GaAs QDs having d_{GaAs} of 30, 20, 15, and 10 nm, respectively.

Figure 1 shows the cross-sectional TEM images of MS InAs/GaAs QDs with d_{GaAs} of 30, 20, 15, and 10 nm which are assigned as Q₃₀, Q₂₀, Q₁₅, and Q₁₀, respectively. In Q₃₀, QDs are formed in each layer; but there is no clear positional correlation. At reduced d_{GaAs} (<20 nm), QDs are stacked and correlated vertically with each other. The vertical correlation of the QDs observed for $d_{\text{GaAs}} < 20$ nm is caused by the strain field of the buried QDs in the underlying layer. The strain that resulted from the underlying layer usually causes atoms to migrate to the top regions of the buried QDs to form QDs in the upper layer.²⁰ It has been reported that although the strain field from a single QD layer penetrates the GaAs spacer layer, it decays rapidly with the distance. In addition, the degree of strain field increases with an increase in the number of layers if d_{GaAs} is smaller than the decay length.⁹ Therefore, a complete vertical stacking of QDs is observed as the d_{GaAs} is reduced.

Figure 2(a) shows the PL spectra at 35 K of Q₃₀, Q₂₀, Q₁₅, and Q₁₀ at an excitation power (P_{exc}) of 100 mW and an excitation energy (E_{exc}) of 3.06 eV. All the PL spectra are comprised of two emission bands. The lower energy side emission is attributed to the ground state transition (GS) and the higher energy side to the emission from the first excited

state (1ES) marked by arrows in Fig. 2(a). Analysis of P_{exc} dependent PL spectra (not shown) confirms that 1ES is the emission from the 1st excited state. The PL spectra are found to be dominated by GS for all d_{GaAs} which is in contrast to the earlier report by Kim *et al.*¹⁷ The peak energies of both GS and 1ES are determined using multi-curve fitting as shown in the inset of Fig. 2(a). The GS peak energy shows a prominent blue-shift of about 129 meV as d_{GaAs} decreases from 30 to 10 nm, i.e., from Q₃₀ to Q₁₀ which is in contrast to the previous reports that show a redshift due to the vertical interaction between the electronic states of the MS QDs.^{8–11} Kim *et al.*¹⁷ have reported the nonmonotonic change of PL peak energy in the MS QDs with a decrease in spacer layer thickness due to the competing effects of increased compressive strain and vertical electronic coupling of the stacked QD layers. The observed blue-shift in Fig. 2(a) with decreasing d_{GaAs} has therefore been attributed to the dominating effect of compressive strain-driven intermixing of In/Ga atoms at the interfaces between InAs QDs and GaAs spacer layers.^{17,21} Energy spacing (ΔE) between GS and 1ES decreases monotonically from 62 to 24 meV as the d_{GaAs} decreases from 30 to 10 nm as shown in inset (a) of Fig. 3. The reduction in ΔE with decreasing d_{GaAs} has been attributed to the compressive strain-driven enhanced interdiffusion of In/Ga atoms at the interfaces between InAs QDs and GaAs spacer layers which is consistent with the observed blue-shift of GS peak energy with decreasing d_{GaAs} [Fig. 2(a)].^{17,21} The PL intensity initially decreases with a decrease in d_{GaAs} , i.e., from Q₃₀ to Q₂₀, and then increases on further decrease in d_{GaAs} , i.e., from Q₂₀ to Q₁₀. The initial decrease in PL intensity from Q₃₀ to Q₂₀ could be attributed to the disorder induced by strain-driven intermixing of the In/Ga atoms at the interfaces between InAs QDs and GaAs spacer layers. The increase in PL intensity from Q₂₀ to Q₁₀ could be associated with the dominating effect of faster carrier relaxation process due to smaller ΔE at reduced d_{GaAs} .

FIG. 2. (a) PL spectra at 35 K of Q₃₀, Q₂₀, Q₁₅, and Q₁₀ at P_{exc} of 100 mW and E_{exc} of 3.06 eV. The arrows indicate 1ES. Inset shows the PL spectrum of Q₁₀ with a multi-curve fitting to GS and 1ES. (b) The E_p dependent TRPL spectra at 35 K of Q₁₀.

FIG. 3. Variation of rise time at GS energy with P_{exc} at E_{exc} of 1.53 and 3.06 eV for Q_{10} as representative of all QDs. The insets (a) and (b) show a variation of ΔE and a rise time at GS energy, respectively, with d_{GaAs} at P_{exc} of 100 mW and E_{exc} of 3.06 eV.

In order to understand the carrier relaxation process, the TRPL spectra at 35 K were recorded at different P_{exc} and E_p at fixed E_{exc} of 1.53 and 3.06 eV. Figure 2(b) shows the TRPL spectra at 35 K of Q_{10} at different E_p , and at fixed E_{exc} of 3.06 eV and P_{exc} of 100 mW as representative of all the samples and excitation conditions. Exponential function was used to determine the rise time by fitting the TRPL spectra. The rise time has been attributed to the carrier capture from the GaAs layers and intra-dot relaxation time within the QDs, and is shown in Fig. 4 at different E_p at fixed E_{exc} of 3.06 eV and P_{exc} of 100 mW. The E_p dependence of rise time represents both the GS and 1ES emission band which can be envisaged from its doublet behavior in Fig. 4. In Q_{30} , the rise time of GS emission energy is longer than that of 1ES. This indicates that the carrier capture rate of 1ES energy level is faster than that of GS. In other words, 1ES energy level is first filled by the carrier capture and then the carriers are relaxed down towards the GS energy level to

occupy through carrier relaxation process. The 1ES energy level therefore acts as intermediate state in the relaxation process in Q_{30} which is in contrast to the earlier report by Bogaart *et al.*¹⁵ They reported that the rise time of the first excited state is longer than that of the ground state in InAs quantum dots, and attributed to the relaxation of carriers through the continuum background, followed by a single LO-phonon emission towards the QD ground state according to the continuum relaxation model. Carrier relaxation via the polaron decay is about 4.5–7 times slower than the observed maximum rise time (~ 10 ps) in Q_{30} .²² The ΔE in Q_{30} is about 62 meV. Thus, the carrier relaxation by a single LO phonon emission is not possible as the LO phonon energy in InAs is ~ 29 meV. Multi-phonon scattering processes²³ and Auger-scattering^{13,14,24} have been proposed to be responsible for the rapid relaxation of carriers in a QD on the time scales of picoseconds. Multi-phonon scattering process is still a too slow process (> 10 ps) and is effective at low excitation level as Auger-like carrier-carrier (cc) scattering process dominates at a high excitation level.^{13,14} The dot density is about $5 \times 10^{10} \text{ cm}^{-2}$, and the carrier density at 100 mW in the first GaAs layer is $\sim 10^{13} \text{ cm}^{-2}$ which is sufficient for the Auger scattering to dominate the carrier relaxation process in Q_{30} . The decrease in PL rise time with increasing P_{exc} is the characteristic feature of the Auger process^{13,14} that can be envisaged from Fig. 3 which further supports the dominance of Auger scattering in carrier relaxation process. Similar behavior is observed in Q_{20} and Q_{15} . However, the rise time of both GS and 1ES in Q_{10} is approximately the same which indicates that the filling rate of carriers in both GS and 1ES energy levels is same which is in contrast to Q_{30} , Q_{20} , and Q_{15} . The ΔE in Q_{10} is 24 meV which is comparable to the LO phonon energy (29 meV) in InAs. Therefore, the carrier relaxation by a single LO phonon emission is possible. The variation of rise time of the GS energy in Q_{10} with P_{exc} is shown in Fig. 3. The rise time is found to be decreased with

FIG. 4. Rise time of Q_{30} , Q_{20} , Q_{15} , and Q_{10} at different E_p at E_{exc} of 3.06 eV (■). The solid curve represents the corresponding PL spectra.

an increase in P_{exc} which indicates that Auger scattering is also responsible for the carrier relaxation process in Q_{10} at $P_{\text{exc}} = 100$ mW in addition to carrier relaxation by single LO phonon emission. The occurrence of carrier relaxation in Q_{10} both by single phonon emission and Auger scattering at 100 mW explains the same filling rate of carriers in GS and 1ES energy levels. Furthermore, the rise time of GS emission energy is found to be decreased with a decrease in d_{GaAs} [inset (b) of Fig. 3]. The decrease in ΔE with a decrease in d_{GaAs} involves less number of phonons for scattering and facilitates the carrier relaxation process by Auger scattering which results in a decrease in rise time with a decrease in d_{GaAs} .

The rise time in Q_{10} is found to be more at $E_{\text{exc}} = 3.06$ eV than that at 1.53 eV which is obvious from Fig. 3. Similar behavior of rise time with E_{exc} is also observed in Q_{30} , Q_{20} , and Q_{15} . This behavior is not associated with the difference in photogenerated carrier density ($\sim 1 \times 10^{13} \text{ cm}^{-2}$ for 1.53 eV and 4×10^{13} for 3.06 eV in the first GaAs layer) since the rise time decreases with an increase in P_{exc} (Fig. 3). As both E_{exc} generates the carriers in the GaAs spacer layer, it is important to consider the band structure of GaAs. The band gap of GaAs is about 1.519 eV, and the bottom of the conduction band is described by the Γ -valley. The L-valley and X-valley are located in the conduction band at about 1.815 and 1.981 eV above the valence band maxima. For 1.53 eV excitation, the carriers are generated in the Γ -valley and then relax down to the bottom of the conduction band of GaAs within the Γ -valley. Subsequently, carriers are captured by the InAs QDs from the GaAs spacer layer. However, for 3.06 eV excitation, the carriers have enough excess energy such that the carriers can be scattered from the Γ -valley to L-valley and X-valley, and return back to Γ -valley by the intervalley scattering process. The difference in rise times at E_{exc} of 1.53 and 3.06 eV varies from ~ 8 to 4.5 ps as the P_{exc} increases. However, the intervalley scattering time in GaAs from Γ - to L-valley is reported to be within 100 fs and that for the L- to Γ -valley is about 2 ps.²⁵ Thus, the observed higher value (4.5 – 8 ps) of difference in rise times at 3.06 eV and 1.53 eV indicates an effective intervalley scattering between Γ - and X-valley, and a slower carrier capture and relaxation at 3.06 eV due to more excess energy. Moreover, the rise time at 1.53 eV decreases at a faster rate than that at 3.06 eV with increasing P_{exc} as shown in Fig. 3 which indicates that Auger scattering is more effective as the E_{exc} is reduced due to less excess energy.

In summary, we have shown the effect of d_{GaAs} on both PL and carrier relaxation process in MS InAs/GaAs QDs. With a decrease in d_{GaAs} , the ΔE decreases which becomes comparable to the LO phonon energy (~ 29 meV) at $d_{\text{GaAs}} = 10$ nm in Q_{10} . Auger scattering is found to be responsible for the carrier relaxation process in the investigated QDs. The carrier relaxation process in Q_{10} is faster in comparison to that in QDs of larger d_{GaAs} (≥ 15 nm) due to the effect of both single LO phonon and Auger scattering. The rise time, i.e., the time for carrier capture and intra-dot

relaxation is found to be longer at $E_{\text{exc}} = 3.06$ eV than that at 1.53 eV due to intervalley scattering process. The lowest carrier capture and intra-dot relaxation time is observed to be ~ 3 ps in Q_{10} at P_{exc} of 100 mW and E_{exc} of 1.53 eV which is much smaller than the value reported earlier in case of single layer QDs.^{12,13} Therefore, multi-stacked InAs/GaAs QDs with complete vertical alignment at reduced d_{GaAs} can be employed for optoelectronic device applications due to the dominating effect of faster intra-dot carrier relaxation process and a higher modal gain.^{6,7}

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